Carrier Dynamics in Monolayer WS_2 via Time-Resolved Terahertz Spectroscopy

Jon K. Gustafson and L. Michael Hayden

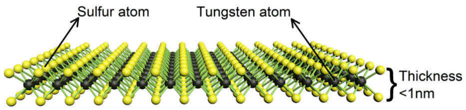
UMBC

Department of Physics, UMBC, 1000 Hilltop Circle, Baltimore, MD 21250

Introduction

Carrier Dynamics: How charges behave in a material under the influence of an electric field

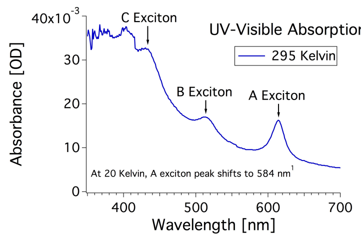
Monolayer: A single layer of molecules

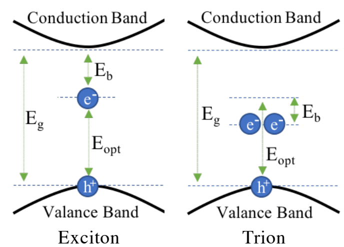


Device Applications: High-speed optoelectronics
Field-effect transistors
Photovoltaics

Properties of WS_2

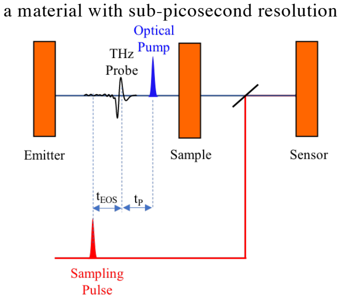
- Reduced dielectric screening results in the existence of tightly bound excitons at room temperature
- Strong Coulombic interactions support charged excitons (trions)

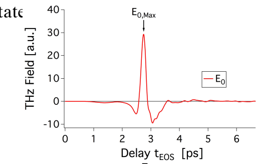


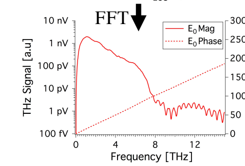


Time-Resolved Terahertz Spectroscopy (TRTS)

- TRTS probes the photo-generated excited state of a material with sub-picosecond resolution

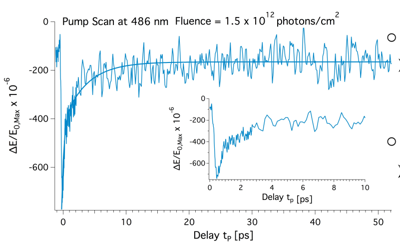






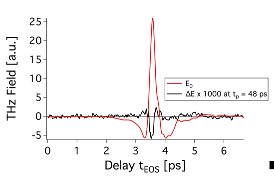
TRTS Modes of Operation

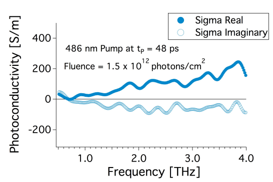
- Pump scan
  - Yields time evolution of photo-carriers
  - $\Delta E/E_0 < 0$ indicates a photo-induced increase in conductivity



- Fast decay ~ 334 fs
  - Unbound electron/hole pairs rapidly forming excitons
- Slow decay ~ 3.7 ps
  - Recombination and exciton-exciton annihilation

- Probe scan
  - Yields complex frequency-dependent conductivity
  - Full THz waveform recorded at various pump delays

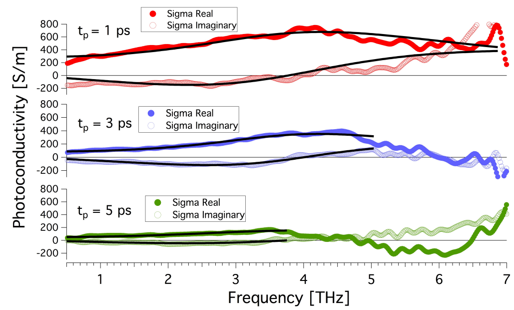




$$\sigma(\omega) = \frac{-(n+1)}{Zd}\frac{\Delta E(\omega)}{E_0(\omega)}$$

Time Evolution of Photoconductivity

- Conductivity was probed at pump delays of t_p = 1, 3, and 5 ps on resonance (584 nm) at 20 Kelvin with ~ 6 x 10^{14} photons/cm^2



- We model the THz photoconductivity as a sum of three oscillators

$$\sigma(\omega) = \sum_{m=1}^{3} \frac{i\, C_m\, \omega}{\omega^2 - \omega_{0m}^2 + i\,\omega\,\gamma_m}$$

C_m ≡ Spectral weight
γ_m ≡ Linewidth (THz)
ω_{0m} ≡ Resonant frequecncy (THz)

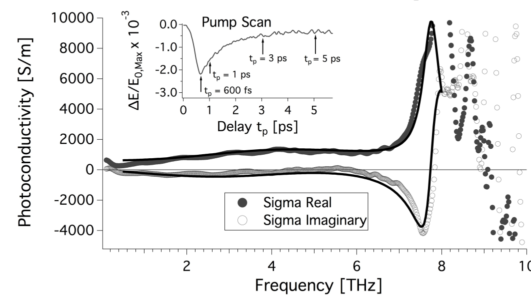
m = 1 → Drude Response m = 2 → Plasma Response m = 3 → Trion Response

Pump Delay	C_1	γ_1	ω_{01}	C_2	γ_2	ω_{02}	C_3	γ_3	ω_{03}
t_p = 600 fs	1.5e16	25.0	0	4.4e16	41.4	4.71	2.64e16	2.93	7.75
t_p = 1 ps	7.8e15	27.0	0	1.9e16	35.1	4.50	0	0	0
t_p = 3 ps	3.9e15	44.9	0	7.5e15	25.8	4.29	0	0	0
t_p = 5 ps	2.1e15	41.1	0	3.3e15	27.3	4.08	0	0	0

- There is no trion component for pump delays of t_p = 1, 3, and 5 ps
- As t_p increases:
  - The ratio C_1/C_2 increases
  - ω_{02} shifts to lower frequencies

Trions

- Trions have been predicted[2,3] and observed[1,4,5,6,7] to have binding energies of about 25-40 meV (~ 6.0-9.7 THz) in WS_2
- The resonant feature in the conductivity at 7.75 THz (32meV) indicates the formation of trions in our sample



Conclusions

- The trend of C_1/C_2 suggests that the Drude response becomes more important relative to the plasma response at longer delay times
- We assign the source of the ω_{02} resonance to a plasmonic[8] response associated with particles with sizes similar to the THz wavelengths
- We see evidence of trion formation at early pump delays

References

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